Photoelectron and Synchrotron Radiation Group(Annual Report)

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Photoelectron and Synchrotron Radiation Group

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Research Activities

I. Studies of optical and electronic properties of solids using synchrotron radiation and related works [1)-5)]

(1) Resonant photoemission in CeNi single crystal.
(T. Kashiwakura, S. Suzuki, T. Okane, S. Sato, T. Kinoshita, A. Kakizaki, T. Ishii, Y. Isikawa, H. Yamagami and A. Hasegawa)

The valence band photoemission of the (010) surfaces of CeNi single crystals has been observed. The features occurring in angle resolved normal emission spectra do not show an appreciable amount of dispersion. The energy distribution curves are squeezed and narrower than those suggested by an existing energy band calculation. Resonant photoemission has been observed both near the $3p$ core election excitation threshold of Ni and $4d$ threshold of Ce. An aspect of the super-

Coster-Kronig(sCK) final state with a post-collision interaction is found near the onset of the resonance in the two-hole bound state satellite. In addition to the two-peak profile in the overall spectral aspect of the $4f$ spectrum, two additional features are resolved. The origin of these feature are discussed. The ratio of Fano's q parameters between the $3p$-$3d$ resonance in Ni ions and the $4d$-$4f$ resonance in Ce ions agrees with a result of a simple theoretical analysis.

(2) Resonant photoemission of rare earth compounds.

(A. Kakizaki, T. Kinoshita, T. Kashiwakura, T. Okane, S. Suzuki, S. Sato, Y. Isikawa, K. Soda, T. Mori and T. Ishii)

The energy distributions of the $4f$ election observed on CeNi, $CeSi_2$, $CeCu_2Si_2$, and RCu_6(R=La, Ce, Pr, Nd, and Sm) by means of the $4d$-$4f$ resonant photoemission are discussed. In $CeCu_6$, the $4f$ spectrum has a line shape similer to the two-peak feature, whereas some additional features are found in single crystalline CeNi and $CeCu_2Si_2$, and , probably, in $CeSi_2$. The origin is tentatively ascribed to the extra screening of the $4f$ states as well as the surface effect. In $PrCu_6$, a few features are clearly discernible but only one broad feature is found in $NdCu_6$. In $SmCu_6$, the mixed valence nature explains the observed spectral features. Qualitative explanations of the $4f$ spectra are made by a localized electron picture.

(3) Evidence for fano interference in spin polarization of the 6-eV satellite in Ni valence-band photoemission.

(T. Kinoshita, T. Ikoma, A. Kakizaki, T. Ishii, J. Fujii, H. Fukutani, K. Shimada, A. Fujimori, T. Okane and S.Sato)

Spin-polarized photoemission spectra have been measured on the ferromagnetic Ni (110) single crystal. Evidence for the Fano interference effect in the spin polarization of the 6-eV satellite of the valence band has been obtained. The results indicate that the interference effect between the direct $3d$ election transition and the $3p$ core electron excitation followed by the MVV super-Coster-Kronig process to form the two-d-hole bound state is important to explain the behavior of the resonant 6-eV satellite. It is also found that the spin polarization is still large under an off-resonant condition, and this indicates that the direct $3d$ transition from the majority-spin state is effective to form the two-hole singlet bound state.

(4) Design and performance of beamline BL-8 at the Photon Factory.

(Y. Hirai, I. Waki, K. Hayakawa, K. Kuroishi, Y. Yasaka, N. Kanaya, Y. Satow and S. Sato)

We designed and constructed beamline BL-8 at the Photon Factory, the National Laboratory for High Energy Physics. The beamline has been in full operation since 1987-1988. On this beamline,one can utilize the synchrotron radiation from a bending magnet in the 40 eV to 35 keV energy range. The beamline has three branch beamlines (8A, 8B, and 8C) where one can perform a wide scope of researchs: e.g., soft X-ray spectroscopy and photochemical reactions on 8A; XAFS on 8B; lithography, microscopy, and micro-tomography on 8C.

(5) X-Ray imaging optics in Japan.

(T. Namioka, K. Yamashita, M. Yamamoto, T. Matsushita, S. Aoki and S. Sato)

Some results obtained as part of a multilaboratory research project entitled "X-Ray Imaging Optics" are presented, with emphasis on a new generation of optical components and systems designed to operate in the soft X-ray (SXR) region. Included are discussions of optical constants for various thin-film materials in the SXR region, SXR multilayers, SXR microscopes, X-ray telescopes, and metrological instrumentation for testing and characterizing SXR optical elements.

II. Studies of solid surfaces and electronic structures of solids [6),7)]

(6) Periodicities and electronic structures of potassium-covered single-domain Si(001) surfaces below saturation coverage.

(Y. Sasaki, Y. Enta, S. Suzuki and S. Kono)

Periodicities of single-domain Si(001) surfaces covered with potassium have been studied by low energy electron diffraction as a function of potassium coverage for ambient (~320 K) and low (100-200 K) substrate temperatures. New phases of potassium-covered Si(001) surfaces have been identified for low substrate temperature. Angle-resolved ultraviolet photoelectron spectra were measured for a mixed phase of $\begin{pmatrix} 2 & 1 \\ 0 & 3 \end{pmatrix}$ and $\begin{pmatrix} 2 & -1 \\ 0 & 3 \end{pmatrix}$, and for a 2×3 phase at low temperature. The surface electronic structure of the former appeared to be metallic and that for the latter semiconducting. The results were discussed based on structural models proposed from XPD [Surf. Sci. 256(1991) 370] for the corresponding phases of Cs-covered Si(001) surfaces with the aid of recent calculations of electronic structures by Morikawa et al. [Phys. Rev. B 44(1991) 3459] .

(7) Photoemission and bremsstrahlung from uranium compounds.

(S. Suzuki, S. Sato, T. Ejima, K. Murata, Y. Kudo, T. Takahashi, T. Komatsubara, N. Sato, M. Kasaya, T. Suzuki, T. Kasuya, S. Suga, H. Matsubara, Y. Saito, A. Kimura, K. Soda, Y. Ōnuki, T. Mori, A. Kakizaki and T. Ishii)

The electronic states of uranium compounds, UGe_2, U_3Ge_4, U_5Ge_3, UC, UB_{12}, U_3P_4, U_3As_4, UPt_2Si_2, U_2PtSi_2, and $U_3T_3X_4$ (T=Ni, Cu; X=Sn, Sb), have been studied by means of X-ray photoelectron spectroscopy and bremsstrahlung isochromat spectroscopy. Ultraviolet photoelectron spectra including those obtained by resonant photoemission are measured on UGe_2, U_3Ge_4, U_5Ge_3. The energy distributions of the 5*f* partical states below the Fermi levels show a common profile that a peak occurs very near to the sharp Fermi edge, the emission intensity decreases smoothly toward the high binding energy and an unresolved feature exists around 2 eV. Bremsstrahlung isochromat spectra have an aspect common to different compounds. Each spectrum has the sharp Fermi edge, a kink or a shoulder at several tenths of an electron volt, and a peak above it. On the high energy side of the peak the intensity decreases gradually and a broad hump around 3.5 eV overlaps with the tail part. The location and magnitude of this hump vary among different compounds. The profiles of the XPS and BIS spectra of UC compared with the energy band DOS curve. Neither the energy band model nor the impurity Anderson model appears to be able to explain the occupied and unoccupied 5*f* spectra of the uranium compounds observed here completely. The U4*f* lines are accompanied by satellites located at about 7 eV from the main peaks and have asymmetric tailing on the high binding energy side of them. The satellite and the tailing are ascribed to a shake down transition. The excitation spectra of the 5*d*-5*f* resonance in U-Ge compounds have well defined two peaks, whereas no line is found in the prethreshold region. This is attributed to the strong spin-orbit interactions of 5*d* electrons.

III. Studies of the electronic structure of high-Tc superconductor and fullerene [8)-19)]

(8) Formation process of fermi-liquid states in high- Tc superconductor studied by photoemission and X-ray absorption.

(T. Takahashi)

The process of formation of the Fermi-liquid states in high-Tc Superconductor

has been studied by angle-resolved photoemission and polarized soft x-ray absorption. The experimental results show that hole-doping in an insulator produces new electronic states in the charge-transfer gap and the density of the new states (mid-gap states) increases with the hole-concentration. It was also found that the mid-gap states are formed by a transfer of weight of the electronic states from the upper Hubbard band and the occupied O2p states. This suggests that the Mott-Hubbard framework of the electronic structure collapses in the superconductor. In the "over-doped" region, on the other hand, a slight rigid shift of the dispersive bands crossing the Fermi level was observed, which suggests that a rigid- band picture is recovered once the Fermi-liquid-like states are formed by the hole-doping.

(9) De Haas-van Alphen measurement of $YBa_2Cu_3O_7$.

(G. Kido, H. Katayama-yoshida and T. Takahashi)

The de Haas-van Alphen effect was measured in oriented powder $YBa_2Cu_3O_7$ using steady high magnetic fields up to 27 T. The oscillatory magnetization was detected by a modulation method. A board band centering at 540±30 T appeared in the Fourier spectrum for H // c-axis, which was attributed to be an extremal cross-section of 0.052 ± 0.003 A^{-2} at the S point. The effective mass was estimated to be 2.1 ±0.5 m_e by the temperature dependence of the osillation amplitude.

(10) dHvA measurements of high-Tc oxides with steady high-magnetic fields.

(G. Kido, H. Katayama-Yoshida and T.Takahashi)

The de Haas-van Alphen (dHvA) effect has been measured in $YBa_2Cu_3O_7$ (Y123) and $Tl_2Ba_2CuO_{6+\delta}$ (T12201) with oriented-powder samples in H // c-axis direction at the temperature range between 1.6 and 4.2 K with steady high magnetic fields up to 27 T. The oscillatory magnetization was detected by a modulation technique. We performed spectral analysis by employing the maximum entropy method (MEM). In Y123, spectral density shows a peak at 456 T which corresponds to an extremal cross-section of 0.044 A^2. The dingle temperature was evaluated to be 7 K using the effective-mass-value of 2.1m_e. In T12201, the maximum of spectral density appeared at 400 T not only for superconducting sample ($\delta \sim .08$) but also for metallic non-superconducting sample ($\delta = .1$).

(11) Photoemission study of the electronic structure of C_{60} and C_{70} and their alkali-metal compounds.

(T. Takahashi)

The electronic structure of solid C_{60} and C_{70} and their alkali-metal (K and Rb) compounds have been studied by ultraviolet photoemission spectroscopy. A series of photoemission spectra measured at each stage of the alkali-metal doping show a unique variation against the alkali-metal content, which is not explained in terms of a simple band-filling picture with alkali s electrons. It was found that when the compound shows the minimum resistivity and as a result the highest superconducting transition temperature, the density of states at the Fermi level shows the maximum. This favors a BCS-like mechanism for the superconductivity in this system. A remarkable difference in the photoemission spectrum was observed between C_{60} and C_{70} when they are doped with the alkali-metal.

(12) Photoemission and inverse photoemission study of alkali-doped C_{60}.

(T. Takahashi, T. Morikawa, H. Katayama-Yoshida, S. Hasegawa and H. Inokuchi)

Comparative photoemission and inverse photoemission measurements were performed on pristine C_{60} and C_{70} and their alkali (K, Rb, Cs) doped compounds to study the electronic structure, in particular its change upon alkali-doping, and the difference between the superconductive compound (K or Rb-C_{60}) and the non-superconductive ones (Cs-C_{60}, Rb-C_{70}).

The electrical conductivity of the sample films was simultaneously mesured *in-situ* to study the correlation between the electrical conductivity and the photoemission spectrum.

(13) Pseudo-gap and electronic structure near the Fermi level in doped C_{60}.

(T. Takahashi)

This Comment discusses the electronic structure of alkali-doped solid C_{60} based on the results of reccent photoemission and inverse photoemission spectroscopies, which have revealed the existence of a pseudo-gap at the Fermi level at the composition of A_3C_{60} (A: alkali metal). Some possible origins for the pseudo-gap are discussed in connection with the superconducting mechanism. Comparison with cuprate superconductors is also made to elucidate the similarity and dissimilarity of the electronic structure near the Fermi level.

(14) Photoemission and inverse photoemission of alkali-doped C_{60}.

(T. Takahashi, T. Morikawa, H. Katayama-Yoshida, S. Hasegawa and H. Inokuchi)

Photoemission and inverse photoemission measurements were performed on alkali-doped C_{60}. It was found that alkali-doping in solid C_{60} does not cause a rigid shift of Fermi level as in a rigid band model, but cause splitting of the LUMO band across the Fermi level, resulting in formation of an energy gap at the Fermi level at the composition of A_3C_{60} (A; alkali metal). Strong electron correlation and lattice distortion (Jahn-Teller effect) were discussed as possible origins for the pseudo-gap. It was also found that the density of states at the Fermi level in nonsuperconducting compounds (Na-C_{60}, Cs-C_{60}, Rb-C_{70}) is much smaller than those of superconducting compounds (K-C_{60}, Rb-C_{60}). Comparison of photoemission and inverse photoemission spectra with cuprate high-Tc superconductors were also made.

(15) Ultraviolet photoelectron spectra of C_{76} and K_xC_{76}.

(S. Hino, K. Matsumoto, S. Hasegawa, H. Inokuchi, T. Morikawa, T. Takahashi, K. Seki ,K. Kikuchi, S. Suzuki, I. Ikemoto and Y. Achiba)

Ultraviolet photoelectron spectra of C_{76}, one of the fullerene family of compounds, have been measured with a synchrotron radiation light source. The photoemission spectral onset is 1.3 eV below the Fermi level, which is equal to that of C_{84} but is about 0.5 eV smaller than that of C_{60} or C_{70}. The relative intensity of the photoemission spectral features is independent of the incident photon energy change. Photoemission spectral changes for C_{76} during potassium dosing are also measured. A new band becomes distinct between the Fermi level and the HOMO band, when the potassium content (x in K_xC_{76}) is equal to or greater than 1.1. The spectral onset moves toward the Fermi level with increasing potassium dosage, approaching it but never crossing it. This indicates that potassium-dosed C_{76} could be a narrow gap semiconductor.

(16) Review of photoemission studies of super conductive alkali-doped C_{60}. [16)-19)]

T. Takahashi

Photoemission study on superconductive alkali-doped C_{60} was reviewed. It has been found by photoemission spectroscopy that alkali-doping into solid C_{60} does not necessarily cause a rigid band-filling but rather produces new electronic states near the Fermi level. This peculiarity in the eleconic structure in alkali-doped C_{60} may be

due to an effect of a relatively strong electron correlation in molecular C_{60} solid. Some possible mechanisms for the superconductivity are discussed.

Doctor Thesis (March 1993)

(1) Natsuo Nakamura
Development of an apparatus for surface-structure-analysis based on micro-beam and study of in adsorbed Si(111) surfaces

Master Thesis (March 1993)

(1) Yuji Kudo
X-ray photoelectron and bremsstrahlung spectroscopies of cerium compounds.
(2) Hideyuki Takahashi
X-Ray photoelectron Diffraction study of noble-metal adsorbed W(100) surfaces
(3) Takashi Morikawa
Photoemission study of alkali-doped C_{60} compounds.

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